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(6) INTERPRETATION OF EXPERIMENTAL MOSSBAUER SPECTRUM AREAS.

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INTERPRETATION OF EXPERIMENTAL MOSSBAUER SPECTRUM AREAS †

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ABSTRACT

A ~~The~~ method ~~of Shirley and coworkers~~[1] for interpreting Mossbauer spectrum areas is reformulated and extended to more general line shapes. The treatment is independent of source line shape and may be applied to multiple line absorbers, provided that the contribution to the spectrum of a given absorber line is distinct.

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I. INTRODUCTION

In Mossbauer effect experiments it is frequently of interest to determine absorption cross sections or relative cross sections. Because of saturation effects these quantities are not proportional to observed peak heights or areas except in the limit of thin absorbers. The spectrum line areas are of particular interest because they are independent of source line shape and instrumental velocity resolution and because they saturate less rapidly with increasing absorber thickness than do the associated heights.

In the following discussions it will be assumed that irrelevant background counting rates have been subtracted, so that the corrected experimental counting rate corresponds to gamma rays which arise from the isomeric transition of interest and pass through the absorber before being detected. The words "absorber line" will refer to the energy dependent cross section for absorption, as distinguished from the experimentally observed spectrum line. The width of a line will be the full width at half maximum, irrespective of shape.

II. ANALYSIS

The source is assumed to emit radiation from the transition of interest into the solid angle subtended by the detector at a rate S_0. A fraction f of this rate results from recoilless emissions, and is distributed in energy according to the function $S(E)$, where

$$\int_0^\infty S(E)\, dE = f S_0$$

The quantity f refers to the radiation which escapes from the source; is not the Lamb-Mossbauer factor if the source exhibits resonant self-absorption.

In the absence of extranuclear effects, the absorber nucleus would have an energy dependent cross section for resonant absorption given by

$$\sigma(E) = 2\pi \lambda\!\!\!^{-2} \frac{(2I_e+1)}{(2I_g+1)} \frac{1}{(1+\alpha)} \frac{(\Gamma_0/2)^2}{(E-E_e)^2+(\Gamma_0/2)^2} = \sigma_0 \frac{(\Gamma_0/2)^2}{(E-E_e)^2+(\Gamma_0/2)^2}$$

where $\lambda\!\!\!^{-}$ is the reduced wave length of the resonant radiation, I_e is the excited state nuclear spin, I_g is the ground state nuclear spin, α is the internal conversion coefficient, E_e is the excited state energy, and Γ_0 is the natural level width of the excited state. When internal fields are present in the absorber, the absorption line may split into several components. In addition, temporal or spatial inhomogeneities of the internal fields may cause apparent broadening of these component lines. The effective nuclear resonant cross section for gamma rays of energy E would then be given by

$$\sigma(E) = \sigma_0 \frac{\pi}{2} \Gamma_0 K(E)$$

where

$$\int_0^\infty K(E)\, dE = 1$$

The energy dependent part, $K(E)$, is regarded as being composed of q lines, such that

$$K(E) = \sum_{i=1}^{q} b_i k_i(E)$$

For convenience, the $k_i(E)$ are normalized to a maximum value of unity.

Initially, two mathematically amenable expressions for $k_i(E)$ will be considered. For the Lorentzian case we have

$$k_i(E) = (\Gamma_i/2)^2 \left[(E-E_i)^2 + (\Gamma_i/2)^2\right]^{-1}$$

This would be applicable in the absence of field inhomogeneity if Γ_i is taken to be equal to Γ_o. The Gaussian case, which might be applicable in the presence of extreme field inhomogeneity is given by

$$k_i(E) = \exp\left[-(2/\Gamma_i)^2 (E-E_i)^2 \ln 2\right]$$

The part of the absorption cross section associated with ith line is then

$$\sigma_i(E) = \sigma_o \frac{\pi}{2} \Gamma_o \, b_i \, k_i(E)$$

When the source gamma ray energy is augmented by an amount $\mathcal{E}$ (by Doppler shift), photons are detected at a rate given by

$$R(\mathcal{E}) = (1-f) S_o e^{-\sigma_e m} + \int_{\mathcal{E}}^{\infty} dE \, S(E-\mathcal{E}) e^{-\sigma_e m} e^{-\sigma(E) f' w}$$

where σ_e is the average atomic cross section for non-resonant absorption, m is the surface density of atoms in the absorber, f' is the absorber recoilless fraction, and w is the surface density of atoms capable of resonant absorption. It is convenient to consider the normalized counting rate, $N(\mathcal{E})$, which is unity at high energy shift. $N(\mathcal{E})$ is given by

$$N(\mathcal{E}) = \frac{R(\mathcal{E})}{R(\infty)} = (1-f) + \frac{1}{S_o} \int_{-\infty}^{\infty} dE \; S(E-\mathcal{E}) e^{-\sigma(E) f' w}$$

where the narrowness of $S(E)$ is the justification for the extension of the lower limit to $-\infty$.

The area of the normalized spectrum is given by

$$A = \int_{-\infty}^{\infty} [1 - N(\mathcal{E})] d\mathcal{E} = \frac{1}{S_o} \int_{-\infty}^{\infty} d\mathcal{E} \int_{-\infty}^{\infty} dE \, S(E-\mathcal{E}) \left[1 - e^{-\sigma(E) f' w}\right]$$

$$= f \int_{-\infty}^{\infty} dE \left[1 - e^{-\sigma(E) f' w}\right],$$

an expression which is independent of source strength and line shape, electronic absorption, detector efficiency and solid angle, and instrumental velocity resolution. The normalized spectrum area, A, has the dimensions of energy. Fig. 1 shows a typical spectrum and illustrates the quantities used here. The part of A which is attributable to the ith absorption line is obtained by replacing $\sigma(E)$ with $\sigma_i(E)$.
It may be written

$$A_i = f \int_{-\infty}^{\infty} dE \left(1 - e^{-t_i k_i(E)}\right)$$

where

$$t_i = \sigma_0 \frac{\pi}{2} \Gamma_0 b_i f' w$$

The quantity t_i is the absorber thickness in natural units for the peak of the ith absorption line. The exponential in the expression may be expanded in a power series and, for known $k_i(E)$, the integration may be performed. For the cases considered we have

$$\text{(2a)} \quad A_i = f \sum_{p=1}^{\infty} \frac{(-1)^{p+1}}{p!} t_i^p \int_{-\infty}^{\infty} [k_i(E)]^p dE$$

$$\text{(2b)} \quad = f \Gamma_i \frac{\pi}{2} \sum_{p=1}^{\infty} \frac{(-1)^{p+1}}{p!} \frac{(2p-3)!!}{(2p-2)!!} t_i^p = f \Gamma_i \frac{\pi}{2} L(t_i) \text{ (Lorentzian)}$$

$$\text{(2c)} \quad = f \Gamma_i \frac{1}{2}\sqrt{\frac{\pi}{\ln 2}} \sum_{p=1}^{\infty} (-1)^{p+1} \frac{t_i^p}{p!\sqrt{p}} = f \Gamma_i \frac{1}{2}\sqrt{\frac{\pi}{\ln 2}}\, G(t_i) \text{ (Gaussian)}$$

The saturation functions $L(t_i)$ and $G(t_i)$ are listed in table I and plotted in figure 2. Notice that both have been defined so as to have

unit slope at zero thickness. The Lorentzian saturates less rapidly than the Gaussian because its broad wings account for a larger fraction of its area. The shape of a real absorption line will result from the folding of the mean life dependent Lorentzian into a function which results from extra-nuclear (and usually statistical) processes. ~~If the~~ latter function is Gaussian, the net profile will have a saturation characteristic which lies somewhere between $L(t_i)$ and $G(t_i)$. In order to illustrate this, the case in which $k_i(E)$ results from the folding of a Lorentzian of width Γ' into a Gaussian of width $\delta\Gamma'$ has been calculated for δ equal to $\frac{1}{2}$, 1, 2 and 4. Expression (2a) becomes

$$A_i = f\,\Gamma' C_\delta M_\delta(t_i)$$

where

$$C_\delta = 1.795,\ 2.226,\ 3.219,\ 5.306; \qquad \delta = \tfrac{1}{2},\ 1,\ 2,\ 4.$$

Note that Γ' in the expression above is the width of the Lorentzian component of the absorber line. The total widths are larger by a factor of 1.218, 1.638, 2.587, and 4.560, for δ equal to $\frac{1}{2}$, 1, 2, and 4 respectively. The saturation functions $M_\delta(t_i)$ are listed in Table I and plotted in figure 2. They are defined so as to have unit initial slope.

Because the situation described in the paragraph above is a fairly common one, $W_\delta(t_i)$, the width of the observed spectrum line has been calculated. The calculation assumes no resonant absorption in the source and a Lorentzian emission line of width Γ' (i.e. the same width as the Lorentzian component of the absorption line). The curves $W_\delta(t_i)$ are shown

in figure 3. For the case of an absorber with natural line shape, see the lowest curve of figure 3 of the paper of Margulies and Ehrman[2].

III. SOME APPLICATIONS

A common situation to which (2a) is applicable is the following: The absorption spectrum contains well resolved lines as a result of magnetic dipole or electric quadrupole interaction and it is desired to determine the relative integrated cross sections. Considering only two absorption lines for simplicity, and assuming Lorentzian shape, the desired quantity is $\Gamma_i t_i / \Gamma_j t_j$. The spectrum area A_i is proportional to $L(t_i)$, where $L(t_i)$ is always less than t_i. If a second spectrum is determined using a similar absorber β times as thick as the original one, the ratio

$$\frac{A_i}{A_i^{\circ}} = \frac{L(t_i)}{L(\beta t_i)}$$

is determined. This uniquely specifies t_i and $L(t_i)$. The fitting may be visualized as the process of placing on the $\ln L(t_i)$ vs $\ln(t_i)$ curve a chord of given length and slope. This process is repeated for the second line and the desired quantity is given by

$$(3) \quad \Gamma_i t_i / \Gamma_j t_j = \frac{A_i t_i}{L(t_i)} \Big/ \frac{A_j t_j}{L(t_j)}$$

Notice that the right hand side of (3) does not depend explicitly upon f, Γ_i, or Γ_j, but it does depend upon the absorption line shape which is assumed.

As a check on the assumed absorber line shape, a third measurement may be performed with a similar absorber κ times as thick as the first. Two chords of known length and slope with a common end point must now be fitted to the ln $L(t_1)$ vs ln (t_1) curve. This will hot be possible if the wrong shape has been assumed.

The use of (2) to determine f when Γ_λ is known and vice versa is obvious and will not be discussed.

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1. D. A. Shirley, M. Kaplan, and P. Axel, Phys. Rev. **123**, 816 (1961).

2. S. Margulies and J. R. Ehrman, Nuclear Instruments and Methods **12**, 131 (1961).

t_1	L	$M_{1/2}$	M_1	M_2	M_4	G
0.0	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.2	0.1905	0.1899	0.1892	0.1884	0.1877	0.1866
0.4	0.3637	0.3616	0.3590	0.3558	0.3531	0.3491
0.6	0.522	0.518	0.512	0.505	0.500	0.491
0.8	0.667	0.660	0.651	0.640	0.630	0.616
1.0	0.801	0.791	0.777	0.761	0.746	0.725
1.2	0.926	0.911	0.893	0.870	0.851	0.822
1.4	1.042	1.023	1.000	0.970	0.945	0.907
2.0	1.347	1.316	1.275	1.224	1.180	1.113
2.4	1.524	1.484	1.430	1.364	1.306	1.218
3.0	1.759	1.705	1.633	1.543	1.463	1.341
4.0	2.095	2.019	1.916	1.785	1.669	1.489
5.0	2.383	2.286	2.155	1.985	1.833	1.594
6.0	2.639	2.523	2.365	2.157	1.970	1.673

Table I

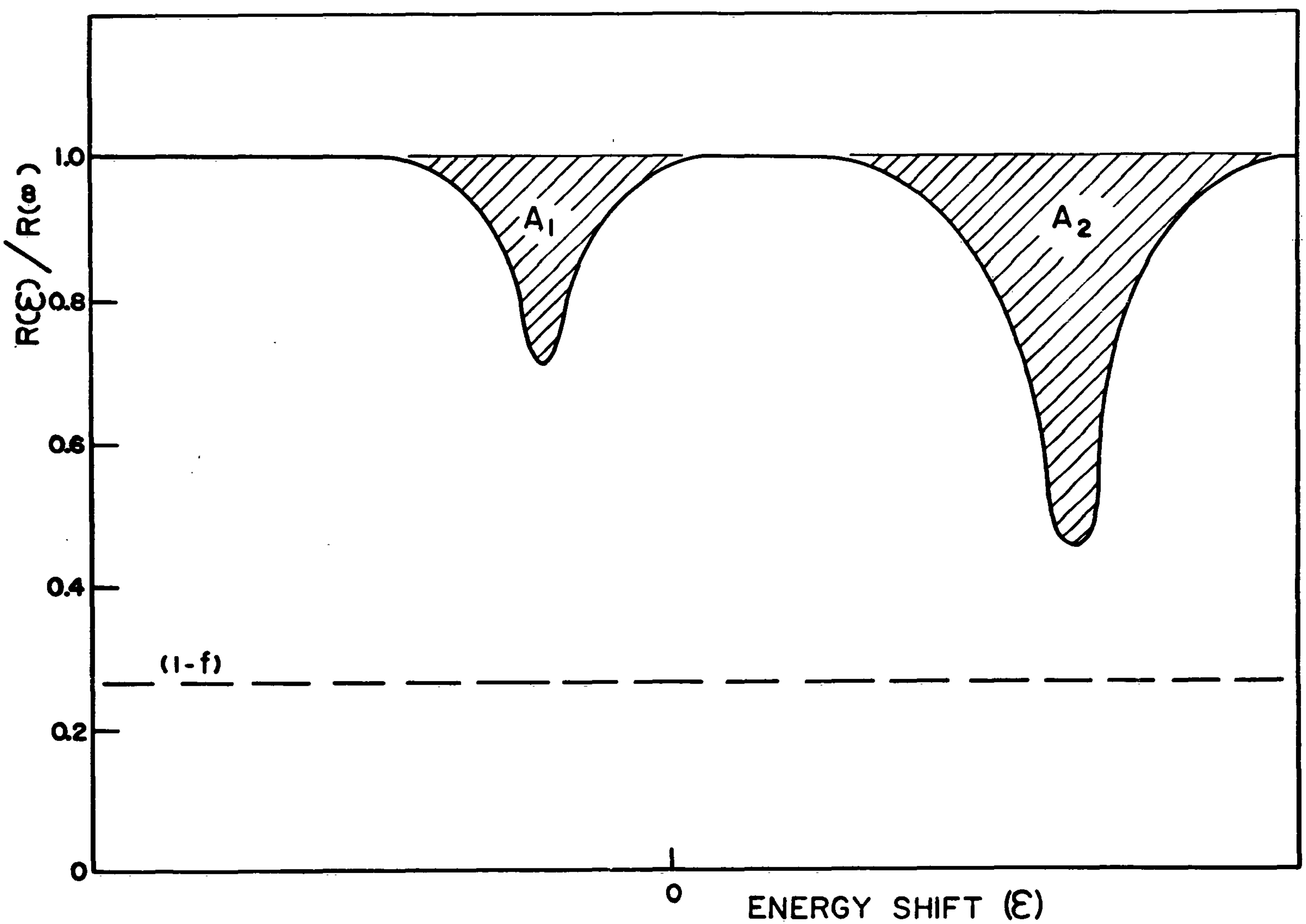
Saturation functions relating normalized spectrum area to absorber thickness for various assumed absorber line shapes.

FIGURE CAPTIONS

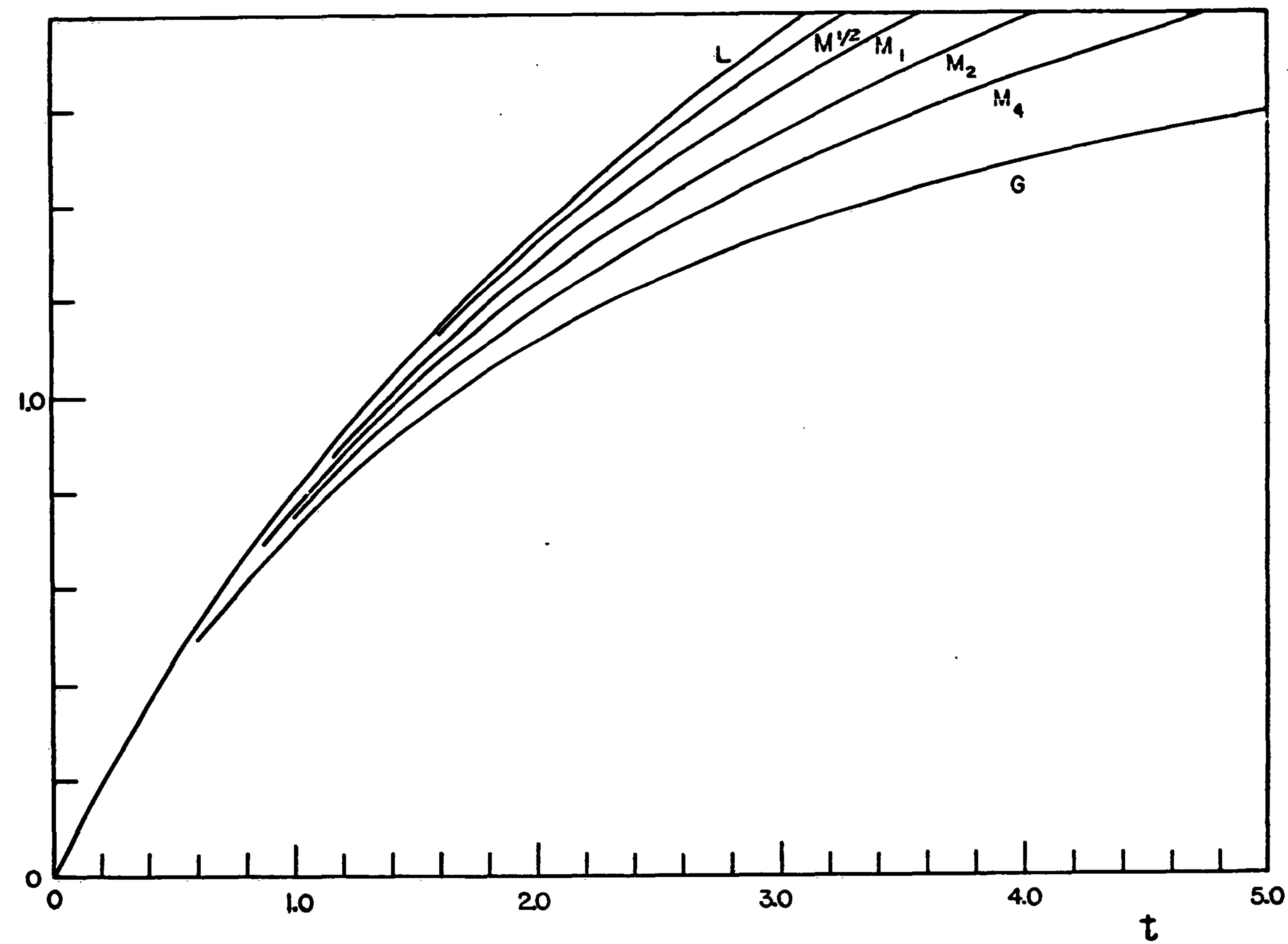
Fig. 1 A typical experimental Mossbauer spectrum showing relative transmission as a function of energy shift for a two line absorber and unsplit source.

Fig. 2. Saturation functions, showing the dependence of normalized spectrum line area upon absorber thickness for various assumed absorber line shapes. Lorentzian, Gaussian, and intermediate shapes are considered. Thickness is measured in natural units, i.e., $t = 1$ corresponds to one absorption length for a monoenergetic source beam on resonance.

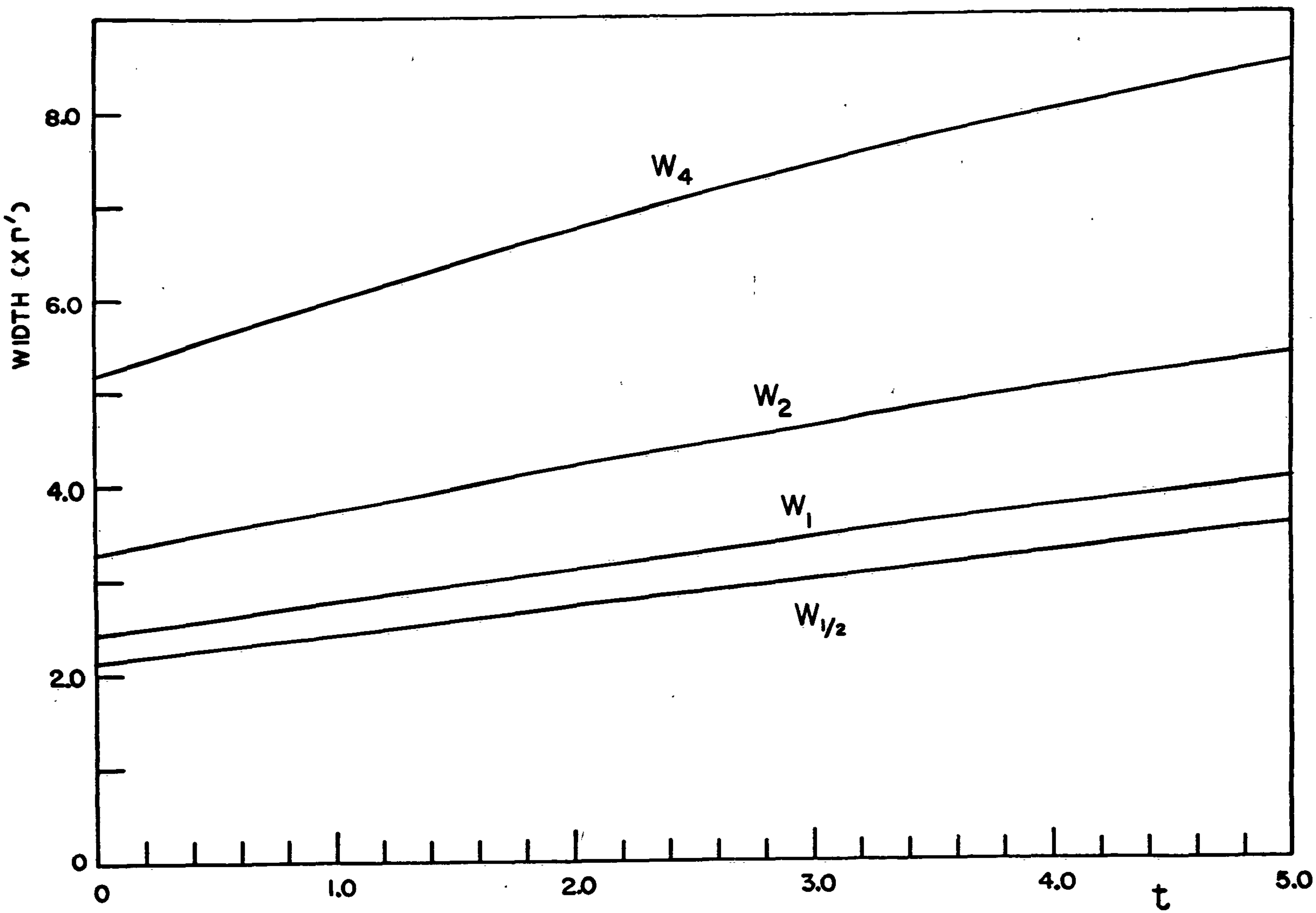
Fig. 3 Width of the observed spectrum line as a function of absorber thickness. The emission line of the source is assumed Lorentzian with a width Γ'. The assumed absorber line shapes result from the folding of a Lorentzian of width Γ' with a Gaussian $\frac{1}{2}$, 1, 2, and 4 times as wide.



G. Lang Fig. 1



G.Lang Fig 2



G. Lang Fig. 3